EFFECT OF FILLER'S TYPE, CONTENT AND MIXING METHOD ON PROPERTIES OF LOW DENSITY POLYETHYLENE/NATURAL RUBBER (LDPE/NR) COMPOSITES

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بسم الله الرحمن الرحيم

In the name of Allah, the Beneficent, the Merciful

With blessings and peace be upon the most honorable Prophets and Messengers, Muhammad and his Folk, Companions and those who follow noble way.

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KESAN TERHADAP JENIS DAN KANDUNGAN PENGISI SERTA KAEDAH PENCAMPURAN SIFAT-SIFAT KOMPOSIT POLIETILENA BERKETUMPATAN RENDAH/GETAH ASLI

Abstrak

Kajian ini berasaskan penyediaan komposit polietilena berketumpatan rendah (LDPE) dan getah asli (NR) yang mengandungi tiga jenis bahan pengisi (silika, kanji jagung dan hampas kelapa) yang mengandungi pelbagai nisbah dan menggunakan kaedah pencampuran yang berbeza. Kaedah pencampuran bahan pengisi ini melibatkan LDPE/NR (campuran I), LDPE (campuran II) dan NR (campuran III). Kesemua sebatian ini disediakan menggunakan kaedah pencampuran tertutup Brabender pada suhu 150 ^{o}C selama 10 minit. Campuran 1 melibatkan LDPE yang dicampur bersama NR dan kemudiannya bahan pengisi yang lain. Campuran II, LDPE dicampur bersama bahan pengisi dan kemudiannya bersama NR. Manakala campuran III, bahan pengisi dicampur bersama NR menggunakan mesin dua penggelek, setelah itu menggunakan pencampuran tertutup untuk dicampurkan bersama LDPE. Keputusan menunjukkan bahawa, kekuatan tegangan yang diperolehi dari campuran I dan II menunjukkan peningkatan apabila bahan pengisi silika semakin bertambah. Namun kekuatan tegangan menurun apabila pertambahan bahan pengisi silika ditambahkan dalam pencampuran ke III. Manakala pada sebatian silika di campuran II mencapai kekuatan tegangan yang tinggi. Kekuatan tegangan untuk sebatian LDPE/NR bersama bahan pengisi organik (kanji jagung-hampas kelapa) untuk semua kaedah pencampuran menunjukkan penurunan apabila nisbah pengisi bertambah namun demikian, kekuatan tegangan pada ketika kanji jagung ditambah ke dalam kaedah pencampuran II berlaku penurunan dan meningkat setelah nisbah kanji jagung ditingkatkan. Nilai kekuatan tegangan tertinggi untuk sebatian pengisi organik diperoleh dari kaedah pencampuran I. Manakala nilai pemanjangan pada takat putus bagi komposit LDPE/NR dan pengisi diperolehi dari komposit LDPE/NR dan pengisi jenis kanji jagung.

Namun, nilai kekuatan tegangan berfungsi diperolehi daripada komposit LDPE/NR yang mengandungi bahan pengisi jenis silika.

Analisa terhadap keretakan tegangan pada komposit setelah pengekstrakan soxhlet dan keretakan tegangan menunjukkan morfologi bagi NR dan LDPE serta bahan pengisi bergantung kepada jenis bahan pengisi dan kaedah pencampuran yang digunakan. Campuran silika di dalam komposit LDPE/NR menunjukkan fasa NR yang lebih luas berbanding LDPE. Manakala penambahan bahan pengisi kanji jagung ke dalam komposit LDPE/NR menunjukkan kesan penyerakan yang baik tetapi apabila ditambahkan bahan pengisi jenis hampas kelapa ke dalam komposit LDPE/NR menunjukkan kurang berlakunya penyerakkan dan morfologi yang kurang baik berbanding apabila ditambahkan bahan pengisi kanji jagung.

EFFECT OF FILLER'S TYPE, CONTENT AND MIXING METHOD ON PROPERTIES OF LOW DENSITY POLYETHYLENE/NATURAL RUBBER (LDPE/NR) COMPOSITES

ABSTRACT

This study was based on preparation of low density polyethylene (LDPE) and natural rubber (NR) composites with three types of fillers (Silica, Corn Starch and Coconut Husk) and different contents (10, 20 and 30 phr) using three different mixing methods. These methods focused on the mixing order of filler into LDPE/NR (mixing I), in LDPE (mixing II) or in NR (mixing III). All the compounding is carried out by Brabender internal mixer at 150 ^{o}C for 10 minutes. In mixing I, LDPE was mixed with NR followed by addition of filler. Mixing II, LDPE was mixed with filler followed by addition of NR. Mixing III, the filler was mix with NR by two-roll mills and followed with LDPE in internal mixer. Result indicated that the tensile strength in mixing I and II methods was increased with the increasing of inorganic filler (Silica) content, but decreased with increasing content of silica in mixing III method. Silica composite in mixing II achieved the highest tensile strength. At all mixing methods the tensile strength for LDPE/NR composites with the organic fillers (Corn Starch and Coconut Husk) decreased when the filler content increased but the tensile strength with addition of corn starch in mixing II method was decreased and increased when the content of corn starch increased. The highest tensile strength for organic filler composite was obtained from the mixing I method. The highest elongation at break for LDPE/NR composite with the fillers was at LDPE/NR composite with corn starch filler but the highest tensile modulus was at LDPE/NR composite with silica filler.

The tensile fracture surface of the composites and soxhlet extraction showed the morphology of NR and LDPE as well as fillers are strongly dependent on filler types and also mixing methods, where addition of silica to LDPE/NR composite was tended to prefer NR phase more than LDPE phase. Addition of corn starch filler to LDPE/NR composite designated better dispersion but with addition of coconut husk to LDPE/NR composite showed poor dispersion and morphology compared with addition of corn starch filler.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF SYMBOLS AND ABBREVIATION

PE	Polyethylene
NR	Natural Rubber
LDPE	Low Density Polyethylene
HDPE	High Density Polyethylene
LNR	Liquid Natural Rubber
PP	Polypropylene
phr	Part per hundred resin
^{o}C	Degree Centigrade
mm	Millimeter
C	Carbon
H	Hydrogen Atom
CH_3	Methyl
UV	Ultraviolet
TPNR	Thermoplastic Natural Rubber
MRPRA	Malaysian Rubber Producers' Research Association
Si	Silica
C_H	Coconut Husk
C_S	Corn Starch
%	Percentage
%wt	Weight Percentage

MPa	Mega Pascal
g	Gram
cm^3	Cubic Centimeter
µm	Micrometer
min	Minute
KN	Kilo Newton
BET	Brunarer, Emmett and Teller
SEM	Scanning Electron Microscope
ASTM	American Society for Testing Material
m^2	Square meter
MV	Average Viscosity
VM	Minimum Viscosity
M1	Mixing I method
M2	Mixing II method
M3	Mixing III method
M0	LDPE/NR composite without filler
M1Si10	LDPE/NR composite with 10 phr Silica prepared by Mixing I method
M1Si20	LDPE/NR composite with 20 phr Silica prepared by Mixing I method
M1Si30	LDPE/NR composite with 30 phr Silica prepared by Mixing I method
M2Si10	LDPE/NR composite with 10 phr Silica prepared by Mixing II method
M2Si20	LDPE/NR composite with 20 phr Silica prepared by Mixing II method
M2Si30	LDPE/NR composite with 30 phr Silica prepared by Mixing II method

M3Si10	LDPE/NR composite with 10 phr Silica prepared by Mixing III method
M3Si20	LDPE/NR composite with 20 phr Silica prepared by Mixing III method
M1C_S10	LDPE/NR composite with 10 phr Corn Starch prepared by Mixing I method
M1C_S20	LDPE/NR composite with 20 phr Corn Starch prepared by Mixing I method
M1C_S30	LDPE/NR composite with 30 phr Corn Starch prepared by Mixing I method
M2C_S10	LDPE/NR composite with 10 phr Corn Starch prepared by Mixing II method
M2C_S20	LDPE/NR composite with 20 phr Corn Starch prepared by Mixing II method
M2C_S30	LDPE/NR composite with 30 phr Corn Starch prepared by Mixing II method
M3C_S10	LDPE/NR composite with 10 phr Corn Starch prepared by Mixing III method
M3C_S20	LDPE/NR composite with 20 phr Corn Starch prepared by Mixing III method
M1C_H10	LDPE/NR composite with 10 phr Coconut Husk prepared by Mixing I method

$M1C_H20$	LDPE/NR composite with 20 phr Coconut Husk prepared by Mixing I method
$M1C_H30$	LDPE/NR composite with 30 phr Coconut Husk prepared by Mixing I method
$M2C_H10$	LDPE/NR composite with 10 phr Coconut Husk prepared by Mixing II method
$M2C_H20$	LDPE/NR composite with 20 phr Coconut Husk prepared by Mixing II method
$M2C_H30$	LDPE/NR composite with 30 phr Coconut Husk prepared by Mixing II method
$M3C_H10$	LDPE/NR composite with 10 phr Coconut Husk prepared by Mixing III method
$M3C_H20$	LDPE/NR composite with 20 phr Coconut Husk prepared by Mixing III method

CHAPTER 1

INTRODUCTION

1.1 Research Background

The blending of two or more polymers has become an increasingly important technique for improving the cost performance ratio of commercial plastics. For example, blending may be used to reduce the cost of an expensive engineering thermoplastic, to improve the treatments of a high-temperature or heat-sensitive thermoplastic, or to improve the impact resistance. Research on the elastomer-thermoplastic blend has high interested on natural rubber–polyethylene (NR/PE) blends (Choudhury, et al., 1990). The main characteristic of these blends is a two-phase system (Elliot and Tinker, 1990). The first phase is a hard polymer domain which does not melt at room temperature but starts melting at elevated temperatures (LDPE). The second phase is soft rubbery (NR). Structure-wise, polyethylene is expected to be more readily compatible with natural rubber when they are blended (Brydson, 1995). NR/PE blends are normally carried out using a mixer such as a Brabender Plasticorder mixer operating at a rotor speed of between 40 and 60 rpm and a mixing temperature higher than the PE's melting point (Choudhury, et al., 1990). Compatibiliser can also improve the compatibility of phases in natural rubber/polyolefin blends (Abdullah and Ahmad, 1992; Ibrahim and Dahlan, 1998; Dahlan, et al., 2002). It reduces the interfacial tension within the blends, and improves the interfacial adhesion of the blend phases resulting in improved properties (Subramaniam and Mehra, 1987).

Since natural rubber and low density polyethylene are not of the same group of polymer, the expected properties will not be achieved because of poor adhesion between the phases created due to immiscibility in the thermodynamics sense. This situation can be remedied by using certain fillers to improve the interfacial interactions between the phases. One of the elastomer-thermoplastic problems is morphology, and there were some studies to treat that by using Electron-Beam (Dahlan, et al., 2002).

Fillers may be used at concentrations of 10% to 50%, targeted at some desired physical or chemical properties, but also frequently useful as cheapeners. Wherever their major utility is to stiffen and strengthen, they will be termed "reinforcement," and in most cases have a fibrous structure. Particle size and distribution are of highest importance. The inorganic fillers include limestone, quartz, silica, talcum, alumina and other minerals, which are often characterized by its effectiveness and expensive or organic, which is characterized by light weight and affordability, include sawdust, paper, jute or. Currently, low cost fillers are also added to polyolefin and to most polymers, without reducing the profile or impairing melt flow. In order to enhance the adhesion of fillers and polymer matrix an interesting industry of coupling agents has been developed, including stearates, silanes and, recently, titanates. These materials show chemical affinity to both polymer and inorganic filler. In general, fillers improve dimensional stability and impact strength, mainly for brittle thermosets. In addition, there are currently fillers that convert a polymer into an electrical conductor or semiconductor and there are a large collection of synthetic fibers based on polymers (Arie, 1997).

Nowadays, carbon black and silica are two main fillers for rubber reinforcement due to certain of active functional groups on the surface (Yiqing, et al., 2005). Besides, there are locally available materials such as limestone, eggshells, corn cobs, groundnut shells, rubber seed shells, rice husk, cocoa pod husk and cherry are amongst the underutilized renewable resources in our society today which can be used directly or converted by simple processes to valuable materials in polymer or related applications (Adeosun, 2000; Adewisi, 1997; Osabohien, et al., 2004; Ishak and Bakar, 1995; Imanah and Okieimen, 2003; Jideonwo and Utuk, 2000; Ogunniyi, 1989).

1.2 Problem Statement

The morphology study of the polymer particularly elastomer in blend is important to understand. The interface adhesion between natural rubber and low density polyethylene phases that the blend is unpredictable and very hard to dominate. Also the fillers dispersion in LDPE/NR blend is unclear. The components concentration in composite affects composite properties but that in the blends cannot be measured and controlled. Through the above mentioned the properties composites could be difficult to tailor.

This study used elastomer composite that consists of 30 phr NR and 70 phr LDPE with different types of fillers. Those fillers are Silica (inorganic filler), Coconut Husk and Corn Starch (organic). The filler content that was using 10 phr, 20 phr and 30 phr and mixed into LDPE/NR matrix with three mixing methods.